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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air-snowpack exchange of bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS – Part 1: In-snow bromine activation and its impact on ozone" *by* K. Toyota et al.

Anonymous Referee #2

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Toyota et al. describe the development and application of a snow-atmosphere 1-D model used to examine bromine activation in the Arctic springtime. Detailed physical parameters and chemical mechanisms included in the model are described. The development of this model was a massive undertaking, and the authors are applauded for their effort. However, a weakness of the manuscript is that the model assumptions of the bulk snow chemistry are not supported by the scientific literature reporting actual snow chemistry measurements (with the exception of nitrate), as described below. However, the applications of the model to explore the influences of wind

speed (and corresponding boundary layer height), the bromine explosion mechanism, ozone fluxes, and column BrO are very well done and tackle important current questions in Arctic bromine chemistry; these, and the corresponding discussion, are major strengths of the manuscript. Suggested revisions to the manuscript are listed below.

Major comments:

Pg. 20345: It would seem appropriate to also mention recent 1-D modeling by Piot and von Glasow (2008, ACP), who showed ozone depletion within 1 day when recycling on the snow surface was included. The study by Piot and von Glasow represents an intermediate between the Lehrer et al and Thomas et al (and this study).

Sec. 2.2: It is assumed that mercury reactions are included in this model, but that mercury is simply not discussed in this manuscript. Is that correct? Please clarify the text. Also, since several components of the chemical mechanism have changed, it would be useful to include a table in the supplemental information that shows all revisions to the Toyota et al 2004 mechanism. Otherwise, the vague descriptions in Sec. 2.2 do not provide sufficient information for comparison with the chemical mechanisms in other models.

Pg. 20349, Lines 21-23: Please clarify whether HCHO, CH3CHO, and C2H2 were the only hydrocarbons included in the current model.

Pg. 20353, Line 22: Douglas et al 2012 discusses the chemistry of frost flowers. Perhaps a more appropriate reference would be Voisin et al. 2012 (JGR, "Carbonaceous species and humic like substances (HULIS) in Arctic snowpack during OASIS field campaign in Barrow).

Pg. 20353, Sec. 2.6: Please clarify this underlying assumption for this section. Is this saying that soluble species within the LLL on snow grains are physically transferred between snow grains in the snowpack?

Sec. 2.7: This section is extremely long (especially in comparison with the detail in

other sections), and therefore, it may improve readability to perhaps move some of this to the supplementary information. As one example, the discussion of testing various stability functions and the reasoning behind using Cheng and Brutsaert (i.e. much of pg. 20355) could be moved to the supplemental information. Sec. 2.9 is also very long and could be moved partially to the supplemental material.

Table 1: This table may not be critical to the main text and could be moved to the supplementary material. The same could be true for Table 5 (particularly since fluxes are not given for the model-derived species, although this addition would be quite useful). Figures 2b, 3, and 4 could also be moved to the supplemental.

Sec. 2.10: The lengthy discussion of the role of temperature could be condensed significantly, particularly since the main point is that role of temperature is not probed by this model.

Table 3: The Cho et al. and Millero et al. manuscripts do not describe actual measured bulk snow chemistry, and this could be easily confused by a reader. Further, Krnavek et al. 2012 (Atmos. Environ.) provide chemistry data corresponding to nearly 1000 Arctic snow samples, and the median values do not agree with those shown in Table 3, as suggested in the third paragraph on page 20365. Further, the recent work by Pratt et al. (2013, Nat. Geosc.) suggests that the ratio of Br-/Cl- plays a role in Br2 activation; this is also supported by laboratory studies of HOBr uptake and Br2 release (Huff & Abbatt 2002 (J. Phys. Chem. A), Adams et al. 2002 (ACP)). If the snow chemistry values shown in Table 3 are to be used and presented in this model study, then the discussion in Sec. 2.10 should be revised to state that these are not necessarily typical values.

Sections 3.1 and 3.2: These sections are well-written with excellent discussion and represent important contributions to our understanding of bromine chemistry. With that said, the last paragraph in Section 3.2 is not very informative to the main points of the manuscript and is suggested to be moved to the supplemental information since the reason for the chemical solver crash is unknown. This discussion detracts from

the important scientific discussion and results of Section 3.2. The last sentence of the paragraph could simply be moved up and integrated into the first paragraph on page 20375.

Section 3.4: This section contains a significant amount of introductory material that could be moved to the introduction. In fact, the introduction could be revised slightly to provide adequate introduction to the main themes of the results and discussion section; this would also provide the reader with the appropriate context to understand the significance of the model results.

Page 20380, Lines 15-17: It could be confusing to the reader that “deliquesced sea-salt aerosols” are mentioned here (and elsewhere), given that only sulfate aerosols are actually considered in this particular model exercise.

Minor suggestions:

Pg. 20342, Lines 13-17: Long, awkwardly worded sentence.

Page 20347, Line 8: As a comment (but not something that needs to be changed in this manuscript), recent work by Kwok et al. (2011, JGR) provides data from recent IceBridge snow depth studies.

Page 20366: There is discussion of results included here in the methods section, which does not seem appropriate.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 20341, 2013.